

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Naoki NISHIURA <i>et al.</i>	Atty. Docket No.: VX062753 PCT
Application No.: 10/591,326	Art Unit: 1766
Filed: August 31, 2006	Examiner: Shane Fang
Title: ENDLESS TUBULAR POLYIMIDE FILM	Confirmation No.: 9434

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

**RULE 132 DECLARATION**

Dear Sir:

Mr. Naoki NISHIURA hereby states and declares as follows:

1. He graduated from the Department of Polymer Science and Engineering, Faculty of Engineering, Yamagata University, and received a doctorate degree in 2005.

Since 1991, he has been working at Engineering Plastics Division, Gunze Limited, at 1, Toritsuki, Murakuno-Cho, in Konan-Shi, Aichi, Japan, where he has been engaged in research and development on the Advanced Polymer Processing. The following facts are based on his education and experience.

2. He is one of the inventors of U.S. Application No. 10/591,326, filed on August 31, 2006, (hereinafter referred to as the "present application") and is familiar with its contents.

3. At the time of this declaration, elected claims 7, 8, 10, and 12 – 16 are pending in the application, together with withdrawn claims 1 – 6 and 17 – 32. The pending and elected claims 7, 8, 10, and 12 – 16 appear in the Claim Appendix at the end of this declaration.

4. He has read and understood the prior art cited against the claims of the present application in the Office Actions mailed January 4, 2011 and July 27, 2010 in the above-identified application, which prior art was used in rejections of claims 7, 8, 10, and 12 – 16 under 35 U.S.C. §103(a). This prior art included:

Kanetake (US 6,303,054),

Economy (US 4,467,000),

Hasegawa (*Structure and Properties of Novel Asymmetric Biphenyl Type Polyimides* in *Macromolecules*, Vol. 32, No. 2, pp. 387 – 396, 1999), and

Wilson (*Polyimide*, Blackie & Son Ltd., 1990, pp. 1-2, scheme 1.2).

5. In my expert opinion, one of ordinary skill in the art would have not combined the teachings of Kanetake, Economy, Hasegawa, and/or Wilson at the time of the presently claimed invention and arrive at the invention defined in claims 7, 8, 10, and 12 – 16. Specifically, one of ordinary skill in the art would have had no reason to combine Kanetake, Economy, Hasegawa, and/or Wilson at the time of the present invention and arrive at the inventions defined in claims 7, 8, 10, and 12 – 16. Furthermore, one of ordinary skill in the art would have had many reasons for not combining these teachings. My opinions are based on the following facts.

6. *Differences in the Technical Fields*

The subject matter defined in claims 7, 8, 10, and 12 – 16 and the teachings cited against Applicants' claims are directed to different technologies and/or fields of technology, which are identified below. The presently claimed invention is identified as "Material" 1 below. The teachings of Economy are identified as "Material 2" below, and the teachings of Hasegawa, Kanetake, and Wilson are collectively identified as "Material 3" below.

Material 1 – The presently claimed invention is directed to an amic acid oligomer, and a semi-conductive polyimide film produced using the amic acid oligomer.

Material 2 – In contrast to Material 1, the teachings of Economy are directed to a process for coating a substrate with a polyimide by using a composition comprising an amino-terminated amic acid oligomer and a tetracarboxylic acid diester.

Material 3 – The teachings of Hasegawa, Kanetake, and Wilson are directed to a material different from Materials 1 and 2, namely, a polyimide produced using a polyamic acid.

7. *Material 1 – Amic Acid Oligomer, and Polyimide Produced Using the Amic Acid Oligomer*

Unlike the polyamic acid of Material 3, amic acid oligomers are low molecular weight compounds. Therefore, in order to obtain a polyimide film, an "addition-condensation reaction" and an "imidization reaction" (i.e., two reactions) must be suitably performed. For example, if the "imidization reaction" progresses before sufficient progress of the "addition-condensation reaction," the "addition-condensation reaction" will no longer progress. As a result, a compound

of sufficiently high molecular weight cannot be obtained, and a polyimide film having suitable properties cannot be produced.

8. The present inventors solved the above problem, which is specific to amic acid oligomers, by combining specific components (asymmetric and symmetric aromatic carboxylic acid components) at a specific ratio, and providing a semiconductive amic acid composition that can form the desired semiconductive polyimide film. This solution of the present inventors, as identified in the present claims, is not mentioned in any of the teachings of Kanetake, Economy, Hasegawa, and Wilson; and thus would not have been apparent to one of ordinary skill in the art from the combinations of these teachings.

9. *Material 2 – Composition Comprising Amino-Terminated Amic Acid Oligomer and Tetracarboxylic Acid Diester, and Polyimide Film Produced*

The teachings of Economy disclose using a composition comprising an amino-terminated amic acid oligomer and a tetracarboxylic acid diester. In particular, as the tetracarboxylic acid diester, a tetracarboxylic acid diester of a specific alcohol substituted with an electron withdrawing group, such as  $\text{CF}_3\text{CH}_2\text{-OH}$  and  $\text{CH}_3\text{CH}_2\text{-O-CO-CH}_2\text{-OH}$ , is used in the invention of Economy. A polyimide having a high molecular weight is obtained by using the tetracarboxylic acid diester of the specific alcohol. Accordingly, the polyimide of Economy necessarily contains both an amino-terminated amic acid oligomer and a tetracarboxylic acid diester of the specific alcohol, which is significantly and structurally different from the material defined in claims 7, 8, 10, and 12 – 16 of the present application.

10. *Material 3 – Polyimide Produced Using Polyamic Acid*

The polyamic acid solution composition is typically used as a precursor for producing a polyimide film. The polyimide is principally produced by an “imidization reaction.” However, since polyamic acid has a relatively high molecular weight, a polyamic acid solution tends to have increased viscosity. A polyamic acid solution containing carbon black is particularly problematic, as described on page 2, line 18 to page 3, line 11 of the Specification of the present application.

11. An objective of Kanetake is to produce an electrically semiconductive seamless tubular polyimide film by using a semi-conductive polyamic acid composition that has higher storage stability and that maintains a stable electrical resistivity when molded into a molded product. To achieve this objective, *a specific carbon black is added in a specific amount to a polyamic acid.*

12. In contrast, the objective of Economy is to coat a substrate with a polyimide. In order to achieve this objective, Economy uses an amino-terminated amic acid oligomer in combination with a tetracarboxylic acid diester of a specific alcohol substituted with an electron withdrawing group. More specifically, Economy discloses a *composition containing both an amino-terminated amic acid oligomer and a tetracarboxylic diester of a specific alcohol as essential components*, and this composition is reacted to form a polyimide film on a substrate.

13. One of ordinary skill in the art would have understood that the technical fields of the invention of Kanetake and that of Economy are clearly different and one of ordinary skill in the art would not look to one of these technical fields for modification of the other. As stated above in Section 6 entitled "Difference in the Technical Field," the technique of forming a coating film on a substrate by using a composition containing both an amino-terminated amic acid oligomer and a tetracarboxylic acid diester of a specific alcohol substituted with an electron withdrawing group is *completely different* from the technique of forming a tubular polyimide film by using a polyamic acid. At least for these reasons, there would have been no reason for one of ordinary skill in the art to combine the teachings of Kanetake and Economy at the time of the present invention.

13. On the other hand, even if the teachings of Economy were applied to or combined with those of Kanetake, such a combination would only provide a polyimide film of a composition of an amino-terminated amic acid oligomer and a tetracarboxylic acid diester of a specific alcohol substituted with an electron withdrawing group. In the invention of Economy, the tetracarboxylic acid diester of a specific alcohol is a constituent feature indispensable for achieving the objective therein. One of ordinary skill in the art would have not eliminated this constituent or indispensable feature from Economy, because this would make it impossible to achieve the objective of Economy.

14. At least for the facts and reasons identified above, one of ordinary skill in the art would have had no reason or motivation to combine the teachings of Kanetake and Economy and arrive at the inventions defined in claims 7, 8, 10, and 12 – 16, which includes a semiconductive aromatic polyamic acid composition comprising, *inter alia*: an aromatic amic acid oligomer

obtained by reacting an aromatic diamine with an aromatic tetracarboxylic acid component mixture comprising 15 to 55 mol% of an asymmetric aromatic tetracarboxylic acid component and 85 to 45 mol% of a symmetric aromatic tetracarboxylic acid component; carbon black; and an organic polar solvent.

16. He further declares that all statements made here of his own knowledge are true and that all statements are made on information and belief are believe to be true; and further that these statements were made with the knowledge that willful false punishment and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the above-referenced application or any patent issuing therefrom.

March 22, 2011  
Date

Naoki Nishiura  
Naoki NISHIURA

## **CLAIM APPENDIX**

7. A semi-conductive aromatic amic acid composition comprising:
- an aromatic amic acid oligomer only having structural units derived from at least two aromatic tetracarboxylic acid derivatives and an approximately equimolar amount of at least one aromatic diamine;
- carbon black; and
- an organic polar solvent,
- wherein said at least two aromatic tetracarboxylic acid derivatives are a mixture of 15 to 55 mol% of asymmetric aromatic tetracarboxylic dianhydride and 85 to 45 mol% of symmetric aromatic tetracarboxylic dianhydride or a mixture of 15 to 55 mol% of asymmetric aromatic tetracarboxylic acid diester and 85 to 45 mol% of symmetric aromatic tetracarboxylic acid diester.
8. A semi-conductive aromatic amic acid composition according to Claim 7, wherein the aromatic amic acid oligomer is obtained by polycondensation of a mixture of 15 to 55 mol% of asymmetric aromatic tetracarboxylic dianhydride and 85 to 45 mol% of symmetric aromatic tetracarboxylic dianhydride and an approximately equimolar amount of said at least one aromatic diamine in an organic polar solvent at about 80°C or lower.



10. A semi-conductive aromatic amic acid composition according to Claim 7, wherein the aromatic amic acid oligomer is obtained by polycondensation of a mixture of 15 to 55 mol% of asymmetric aromatic tetracarboxylic acid diester and 85 to 45 mol% of symmetric aromatic tetracarboxylic acid diester and an approximately equimolar amount of said at least one aromatic diamine in an organic polar solvent at about 90 to about 120°C.

12. A semi-conductive aromatic amic acid composition according to Claim 7, wherein a number average molecular weight of the aromatic amic acid oligomer is about 1000 to about 7000.

13. A semi-conductive aromatic amic acid composition according to Claim 7, wherein carbon black is present in an amount of about 3 to about 30 parts by weight per 100 parts by weight of a total amount of aromatic tetracarboxylic acid component and organic diamine.

14. A method for producing a semi-conductive seamless tubular polyimide film, comprising: rotationally molding a semi-conductive aromatic amic acid composition according to Claim 7; followed by heating.

15. A semi-conductive seamless tubular polyimide film for use in an intermediate transfer belt in an electrophotographic system produced by a production method according to Claim 14.

16. A method for producing a semi-conductive aromatic amic acid composition comprising:

subjecting at least two aromatic tetracarboxylic acid derivatives and an approximately equimolar amount of at least one aromatic diamine to partial condensation polymerization in an organic polar solvent, thereby yielding an aromatic amic acid oligomer solution only having structural units of the at least two aromatic tetracarboxylic acid derivatives and the at least one aromatic diamine; and

uniformly mixing electrically conductive carbon black powder with the oligomer solution, wherein said at least two aromatic tetracarboxylic acid derivatives are a mixture of 15 to 55 mol% of asymmetric aromatic tetracarboxylic dianhydride and 85 to 45 mol% of symmetric aromatic tetracarboxylic dianhydride or a mixture of 15 to 55 mol% of asymmetric aromatic tetracarboxylic acid diester and 85 to 45 mol% of symmetric aromatic tetracarboxylic acid diester.